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PROGRESS OF SOIL RADIONUCLIDE DISTRIBUTION STUDIES FOR TITLE: NEVADA APPLIED ECOLOGY GROUP - 1981

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University of California

## Progress of Soil Radionuclide Distribution Studies for the Nevada Applied Ecology Group - 1981 by E. H. Essington

Two nuclear sites have been under intensive study by the Nevada Applied Ecology Group (NAEG) during 1980 and 1981, NS201 in area 18 and NS219,221 in area 20. In support of the various studies Los Alamos National Laboratory (Group LS-6) has provided consultation and evaluations relative to radionuclide distributions in soils inundated with radioactive debris from those tests. In addition, a referee effort was also conducted in both analysis of replicate samples and in evaluating various data sets for consistency of results. Los Alamos also contributed to design of a cleanup experiment to test the effectiveness of a truck mounted vacuum device.

This report summarizes results of several of the data sets collected to test certain hypotheses relative to radionuclide distributions and factors affecting calculations of radionuclide inventories and covers the period February 1980 to May 1981.

## NUCLEAR SITE 201

Nuclear Site 201 (NS201) has been under study since 1976 and efforts were directed this year to completion of the inventory and distribution phases of the evaluation. Certain of the studies reported here which support the inventory and distribution measurements include the effects of sieving on radionuclide distributions, distribution of radionuclides in vertical profiles, attempted resolution of uranium isotopes from natural levels, and the estimation of radionuclide inventory in a large mound near the ground zero at NS201.

### A. Sieved Soils

Soil samples collected from NS201 tend to contain considerable amounts of rocky material as shown in Fig. 1. In order to provide the analyst with the least problem in

dissolving soil samples for radiochemical analyses the samples were oven dried, ball-milled, and sieved through a 10 mesh screen (~2 mm opening). The material passing the 10 mesh screen is presumed to contain most of the radioactivity and is committed for analysis. On a series of randomly selected samples the material not passing the 10 mesh screen was analyzed to determine the partition of radionuclides and the amount of radioactivity contributed to the whole sample by the large fraction. Last year results were reported for the gamma emitting radionuclides in those samples; reported here are the results of plutonium analyses.

The data base generated for that test was first evaluated for abnormalities by comparing the <sup>239,240</sup>Pu, determined by chemical separation and alpha spectrometry. with <sup>241</sup>Am, determined by gamma spectrometry, for the coarse fraction and for the fine fraction. Figure 2 shows the relationship between 239,240 Pu and 241 Am in the coarse fraction. The average ratio (ratio of the mean values as suggested by Gilbert) is 9.2. The linear least squares fit of the dare has as its intercept a value close to zero and a good fit indicating direct correlation of 241 Am with 239, 240 Pu in the large fraction. Figure 3 shows a similar treatment for the fine fraction. The ratio of 239. 240Pu to 241Am is 13, somewhat higher than for the coarse fraction. The linear least squares fit of the data intersects the axis nearly at the origin and also indicates direct correlation of the 241 Am with the 239, 240 Pu content. The difference in ratios of 241Am to 239, 240pu in the coarse and fine fractions indicates that a higher proportion of 241 Am relative to 239, 240 pu is present in the coarse fraction suggesting that 241 Am may be locally more mobile (soluble) upon weathering or the fallout particles, weathering that has occurred since July 1962. The average ratios of 239, 240 pu to 241 Am do not compare with ratios reported by Gilbert; the discrepency is being investigated.

The distribution of  $^{239}$ ,  $^{240}$ Pu between the coarse and fine fractions is shown in Fig.4. The data are plotted on a log scale to better show the low activity results and the line Y = 0.01X is drawn for reference only. The ratio calculated from the

means of the two populations indicates that the fines contain about 260 times more 239, 240 Pu than the coarse fraction; for comparison the ratio of 241 Am in the fine VS coarse fractions reported last year is about 170.

Based on those data the act of sieving NS201 soil, after ball milling, would not be expected to seriously influence results if only the fine material is analyzed. Supporting this observation—the fact that in most cases the coarse material accounts for only a small fraction of the total sample.

## B. Large Mound

A large mound of soil and rock debris, shown in Fig. 5, is located close to the ground zero in NS201. An interest was shown by NAEG relative to the amount of radioactive material contained in the mound and its impact on the total site radionuclide inventory. The mound appears to have been created for storage of contaminated material scraped from the ground zero area. Samples of debris were collected from the surface of the mound around the perimeter and top of the mound; the sampling did not include the interior of the mound because of large rocks and cave-ins during attempted sampling of the interior. Figure 6 shows the plan and relief views of the mound and the sampling locations. Use of data from those samples to obtain an inventory is qualitative, however, an indication of the magnitude of the contribution can be estimated. Also, since several of the samples appeared as referee samples a comparison of the data generated by independent laboratories was made.

Separate aliquotes of three samples were analyzed by gamma spectrometry by two laboratories. Comparison of <sup>241</sup>Am determined by the two laboratories is shown in Fig. 7. The mound appears to have been created for storage of contaminanted material scraped from the ground zero area. Considering the historical variations in radionuclide content among aliquots of the same sample the two laboratories are reporting consistent results. A similar comparison of <sup>137</sup>Cs results is shown in Fig. 8. In both the <sup>241</sup>Am

and <sup>137</sup>Cs analyses laboratory number 1 appears to report higher values. However, there are too few results to conclude that one laboratory is reporting higher value results different from the other.

During the time the samples were commissioned for analysis, a serious reduction in funding occurred; many samples originally designated for radiochemistry were diverted to gamma spectrometry only. From those gamma scans several of the more radioactive samples netted results for <sup>239</sup>Pu although with large counting errors. Concurrently the referee had analyzed three of those samples for <sup>239</sup>, <sup>240</sup>Pu by radiochemistry and alpha spectrometry. Those results are shown in Fig. 9. Very good agreement between the two methods was observed suggesting that the gamma sepctrometrically derived values could be used for estimating <sup>239</sup>Pu where sufficient quantities exist.

Based on those tests the amount of the various radionuclides represented by the mound volume was estimated using an average rectangular prism of base 28 x 92 ft and a height of 10 ft. With the assumption that the average radioactivity observed in the samples is representative of the distribution of radioactivity throughout the mound and that the consistency of the mound material is similar to that of the samples, the estimated inventory of the various radionuclides is as shown in Fig. 10. Plutonium and americium represent the largest burden of radioactivity with the presence of detectable quantities of 137Cs, 152Eu, and 60Co.

The above information may be sufficient to set the relative importance of the mound as a source of radioactivity at NS201. Should more definitive inventory and distribution data be needed, however, the mound will have to be completely distrubed to access the interior for samples. Simple coring is not reasonable because of the large rocks, and trenching is not possible because of the looseness of the material and the experience of cave-ins.

### C. Uranium

The original protocol for the study of N5201 called for samples of soil to be analyzed

for uranium; total uranium on a suite of samples from the highest activity stratum and similarly on a suite from the lowest stratum. If sufficient uranium content was detected above natural levels, a series of samples were to be analyzed for isotopic uranium. Samples were not run for total uranium and to date none of the samples has been run for isotopic uranium. However, the samples committed to gamma spectrometry did yield some information for 235U and 2381.

Figure 11 lists those <sup>238</sup>U analyses yielding above-detectable results. All of those samples were collected from within 600 ft of ground zero. Represented are surface samples (0-5 cm), profile samples, and samples from the mound discussed earlier. Uranium-238 in the surface soil was generally quite low. If one assumes that the sample collected from a depth of 20-25 cm, and about 550 ft from ground zero, more nearly represents a natural level of <sup>238</sup>U, then the surface <sup>238</sup>U content is in the order of 2 times that reference level except for the sample from the large mound at ground zero. The vertical distribution of <sup>238</sup>U has not been measured at the site and use of the 20-25 cm sample as a measure of natural 238U at this time is only qualitative. Analyses of samples from other locations on Nevada Test Site and Tonopah Test Range indicate an average  $^{238}\mathrm{U}$ level of 0.001 nCi/g in surface soils. Because of the wide variability in uranium content of soils and minerals on the Nevada Test Site and Tonopan Test Range this value, too, is highly questionable as a reference for NS201. In any event the detectable 238U is within a factor of 2 to 20 times the reference levels and that difference is insufficient to allow easy resolution of device 23SU from natural 23SU at NS201 for purposes of inventory and distribution.

Uranium-235 was also reported from gamma spectrometric analysis of soils from NS201. Those data are summarized in Fig. 12. Uranium-235 levels are plotted against distance from ground zero in order to indicate if a <sup>235</sup>U concentration gradient can be observed in the data set. There appears to be no indication that <sup>235</sup>U levels in the

surface soils are related to distance from ground zero. Also in Fig. 12 are shown the <sup>235</sup>U levels observed in profile samples taken more than 900 ft from ground zero. Uranium-235 in the surface 5 cm averages about 0.5 pCi/g and is not very different from the <sup>235</sup>U observed in the profile samples.

The few <sup>235</sup>U and <sup>238</sup>U observations reported for NS201, based on gamma spectrometry, suggest that uranium that might have originated in the device may not be easily resolvable from natural uranium. Therefore, efforts to complete the original protocol appear not to be warrented.

### D. Soil Profiles

The original protocol for NS201 required the collection of soil profiles from 22 locations in the NS201 fallout area. Last year gamma spectrometric analyses of 10 of the soil profiles collected close to ground zero were reported. Since then the remaining 12 profiles have been collected and analyzed for the major gamma emitters <sup>241</sup>Am, <sup>137</sup>Cs, and radioisotopes of europium. Figure 13 shows the approximate location of the 22 profiles. Data for the most recent collection indicate that most of the gamma emitters are, in many cases, not detectable; resolution of the distribution of radionuclides to any significant depth is currently not attainable. The data do indicate, however, that in most cases more than 10% of the observed radioactivity is located below the 0- to 5-cm depth. Further evaluation will require more detailed analyses involving measurement of <sup>239,240</sup>Pu and perhaps <sup>241</sup>Am by radiochemical separation and alpha spectrometry.

## E. Observations

Relative to completion of inventory and distribution studies at NS201, it appears that certain needs still exist. Radiochemical analysis of soil profiles for <sup>239</sup>, <sup>240</sup>Pu and in some cases <sup>241</sup>Am is indicated. The original protocol specified measurement of <sup>90</sup>Sr, which has not been accomplished either on soil profile samples or on the surface samples collected for inventory purposes. Because of the hazardous nature of <sup>90</sup>Sr, inventory and distribution measurements for that radionuclide are important in completing the evaluation

of NS201. Based on guidance from NAEG a decission should be made as to the need to further evaluate the large mound at the ground zero in NS20!. It appears that efforts to resolve device induced uranium levels in the fallout region of NS201 may not be necessary at this time.

### NUCLEAR SITE 219 and 221

Studies of inventory and distribution at NS219, 221 began with the gridding of the study site, measurment of gross beta, gamma, and alpha surface radiation, collection of special samples, and collection of routine soil and soil profile samples. Figure 14 shows the general features of the study area at NS219, 221. The two explosion craters, the north and south tenced areas constructed for grazing studies, and the general outline of the sampling grid are shown in their relative positions.

## A. Instrumental Survey

Two years ago an instrumental survey was conducted close to the nuclear craters of NS219, 221 and was reported in NVO-181. As part of the soil sampling program additional instrumental readings were taken, which extends the coverage to include the north fenced area some 12 000 ft north of ground zero. The instrumental survey data will not be presented here but will be discussed by Dr. Gilbert and may be presented in the written version of this report. A unique feature of the survey is that the radioactivity levels near each of the two craters is quite different in magnitude. The survey has also been used in selection of a series of soil samples that are in the process of being evaluated for presence identity, and general magnitude of longer-lived radionuclides.

### B. Radionuclide Identification

NAEG has studied in detail several safety shot sites and only one nuclear site (NS201). NS219, 221 appears to be quite different in radionuclide composition relative to either the safety shot sites or to NS201. For those reasons an effort was undertaken to determine the identity of the longer-lived radionuclides that might be present and that could contribute significantly to the dose potential of the area.

7

Two soil samples were collected from outside the 300 ft exclusion zone of each crater and were evaluated using a Ge(Li) detector and gamma spectrometry or using specific radiochemical separations. Figure 15 lists those radionuclides that were easily detectable by gamma spectrometry, or the specific separation unique to plutonium and uranium analyses. A special effort was made to estimate the level of tritium that might still be present in the soils. No effort was made to ascertain the relative levels of the various radionuclides except that 238Pu to 239, 240Pu ratios were considerably higher than has been observed with safety shot samples. Of specific interest is the appearance of considerable 233, 234U. Detection of 125Sb required chemical separation prior to gamma spectrometry suggesting that its concentration was also quite low.

Tritium vwas determined on a separate sample of soil collected in a wash about 4 000 ft north of ground zero. Because the soil was quite dry the tritium was recovered by saturating the soil with distilled water, vacuum distillation of the water, and liquid scintillation counting of the evaporated water. Tritium levels were quite low ranging from 15 to 150 dis/min per g. of soil.

Obviously not all of the radionuclides present could be detected by gamma spectrometry or by the specific radiochemical procedures used. Figure 16 lists several of those radionuclides suspected of being present either as normal fission products, as radioactive decay daughter products, or as created by neutron activation of device components. There may be others but either their production rates or decay half-lives are low and their concentrations in the soils at 13 - 16 years after the explosions would be expected to be low. As noted earlier a series of 20 samples have been selected and committed to detailed analyses in order to quantify the relative amounts of the more important radio-nuclides.

### C. Sieved Soils

The soil materials at NS219, 221 are quite rocky, similar to that observed at NS201. Analysis of soil samples requiring a dissolution procedure could be simplified if first the

sample could be seived to remove the less easily dissolved materials. The soil samples that have been collected were sieved to pass a 10 mesh sieve (~2 mm); in a randomly selected set of 20 samples the >10 mesh and separately <10 mesh fractions were analyzed by gamma spectrometry for those radionuclides that were more easily recognized. Figure 17 represents the comparasion of the radionuclide content of sieved fractions where detectable levels of radioactivity were observed in both fractions.

The data presented in <u>Fig. 17</u> suggest that the distribution of radionuclides among the and fractions may differ with the radionuclide. For example <sup>241</sup>Am appears to distribute more to the fine fraction whereas <sup>152</sup>Eu, <sup>154</sup>Eu, and 102m Rh appears to be more evenly distributed. This observation suggests that a degree of weathering of the original "hot" melt particle may have occurred allowing the more soluble radionuclides to interact with the coarser materials. A note of caution in presenting this hypothesis is that many of the >10 mesh analyses were near the limit of detection of the method; that could have some effect on interpretation of the results. In any event some care should be exercised in accepting results from analyses of the 10 mesh fractions uncorrected for the contribution from the 10 mesh fraction. A larger set of samples should be committed to this type of evaluation to determine that contribution.

## D. 241 Am Counting

Higher levels of <sup>241</sup>Am are normally quantified by gamma spectrometry using a Ge(Li) detector. Some concern has been expressed relative to interferences by other radionuclides in the sample emitting significant energies near 60 the key gamma emenation of <sup>241</sup>Am. At the Los Alamos laboratory such interferences have not been observed for samples from the safety shot sites or from NS201. However, the concern exists for samples from NS219, 221 because of the diverse nature of the contributing radionuclides. It has been estimated that the major radionuclides would be <sup>152</sup>Eu and <sup>154</sup>Eu, which emit the characteristic Gd x-rays upon radioactive decay. In order to test the degree of influence <sup>152</sup>Eu and <sup>154</sup>Eu could have a <sup>241</sup>Am quantification, soil samples were assayed

using two Ge(Li) detectors one designated Nuclear Diodes and the other designated Ortec. The Nuclear Diodes detector was set to encompass the energy region of about 40 to 1500 kev whereas the Ortec system was set to expand the region of 10 to 130 kev. Figure 18 shows portions of the spectra, including the 60 kev <sup>241</sup>Am peak, for each detector. The 60 kev <sup>241</sup>Am peak obtained by the Ortec detector is unaffected by the emissions of 152, 154Eu. Comparison of <sup>241</sup>Am results obtained by both detectors should indicate the degree of interference if any exists for soils from NS219, 221. Figure 19 compares the data derived from the two detection systems of a number of soil samples from NS219, 221. In general no difference was observed as seen by the lineraity of the data. In Fig 20 the effect of <sup>152</sup>Eu on the ratio of <sup>241</sup>Am measured by the two methods is shown. The ratio of <sup>241</sup>Am measured by the two methods is shown. The ratio of <sup>241</sup>Am measured by the systems is plotted against the <sup>152</sup>Eu measured on the Nuclear Diodes system. The linear least squares fit has very little slope suggesting that the ratio and thus the <sup>241</sup>Am measured by the Nuclear Diode System is not influenced by the <sup>152</sup>Eu content of the sample.

Based on this test it appears that <sup>241</sup>Am can be quantified in NS219, 221 soil samples without serious error using the Nuclear Diode Ge(Li) det ction system as has been used in the past by Los Alamos.

## DEEP PROFILES

Several years ago considerable effort was involved in establishing the verticle distribution of plutonium and americium at safety shot sites. The standard procedure established by the original NAEG scientists, evaluated the verticle distribution to a depth of only 25 cm and many of the profiles indicated that small amounts of plutonium had penetrated at least to that depth. A special sampling was initiated to establish the depth to which plutonium could be detected relying on sampling to 50 cm or until hard pan or caliche were intercepted. A series of 20 such profiles were requested apportioned over

the 6 safety shot sites studied by NAEG. Results for three of those profiles are described here. Figure 21 shows the distribution of 239, 240Pu with depth at the GMX site in Frenchman Valley. Detectable levels of 239, 240Pu were observed throughout the profile to a depth of 42.5 cm with significant accumulations at 25- and 35-cm depths. The profile was collected at a location believed to be out of the influence of reworking near ground zero thus the accumulations may be a reflection of differing soil textures or chemical composition with depth.

Figure 22 shows the distribution of 239, 240Pu with depth at the Clean Slate I safety shot site at the Tonopah Test Range. As with the GMX site, plutonium was detected at all depths sampled down to 32.5 cm. There appears to be an increased concentration of plutonium in the deepest fractions suggesting a degree of accumulation. However, samples were not collected from a depth below which plutonium had penetrated thus the maximum depth of penetration nor possible accumulations could be determined.

Figure 23 presents the distribution of 239, 240Pu at the Clean Slate 3 safety shot site also at the Tonopah Test Range. The profile was collected outside the inner fence presumably away from the influence of mechanical reworking. There appear to be accumulations of plutonium at the 7.5-, 17.5-, 22.5-, and 32.5-cm depths and plutonium was detected in all fractions collected.

Based in the three profiles collected and analyzed the maximum depth of penetration of plutonium has not been ascertained; further, sampling and analysis would be required to complete the evaluation.

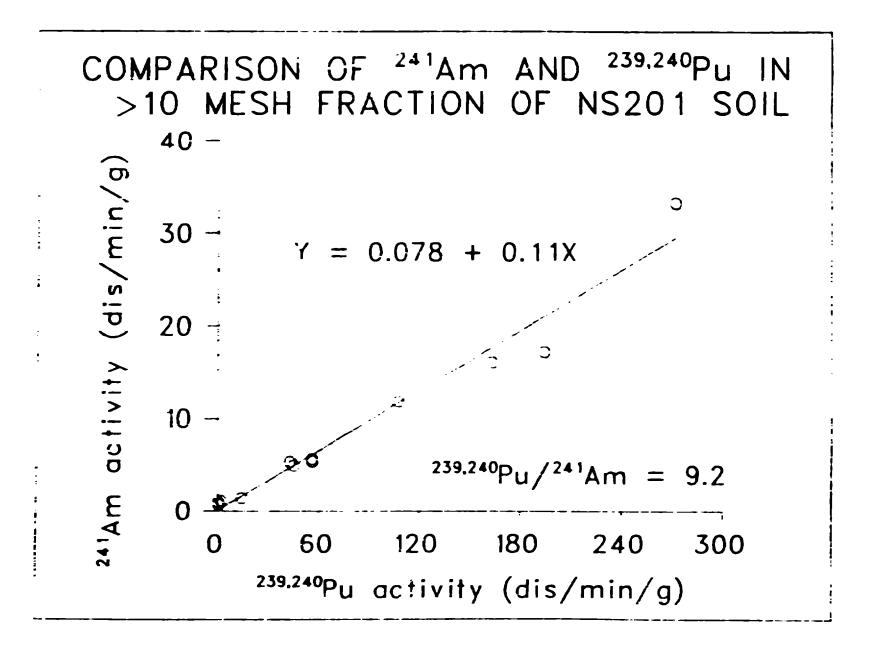
What has been presented here is a brief description of the various data sets acromulated this past year relative to inventory and distribution of radionuclides at Nevada Test. Site and environs. They have indicated areas where more work might be fruitful.

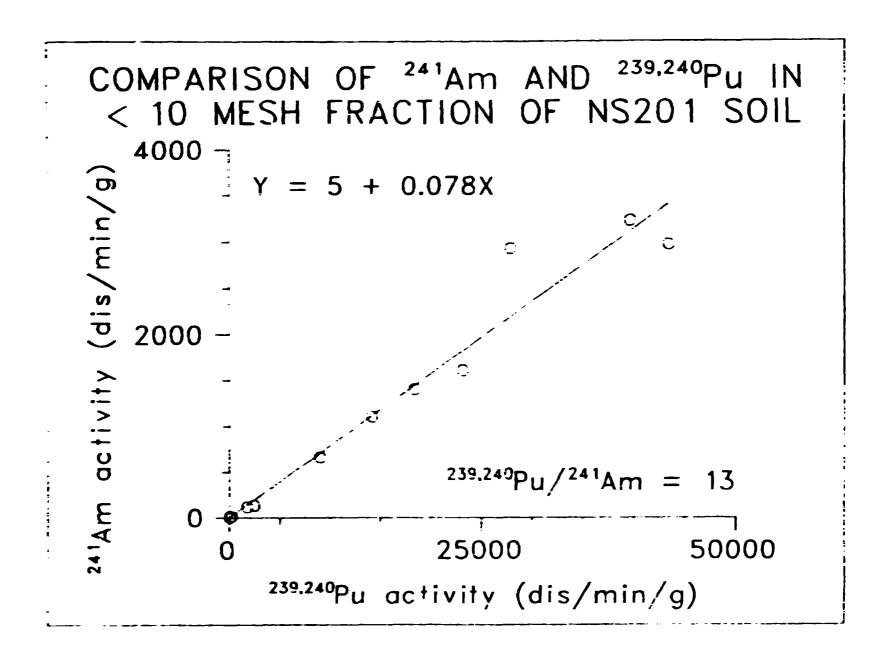
Figure 22 shows the distribution of <sup>239</sup>, <sup>240</sup>Pu with depth at the Clean Slate 1 safety shot site at the Tonopah Test Range. As with the GMX site, plutonium was detected at all depths sampled down to 32.5 cm. There appears to be an increased concentration of plutonium in the deepest fractions suggesting a degree of accumulation. However, samples were not collected from a depth below which plutonium had penetrated thus the maximum depth of penetration nor possible accumulations could be determined.

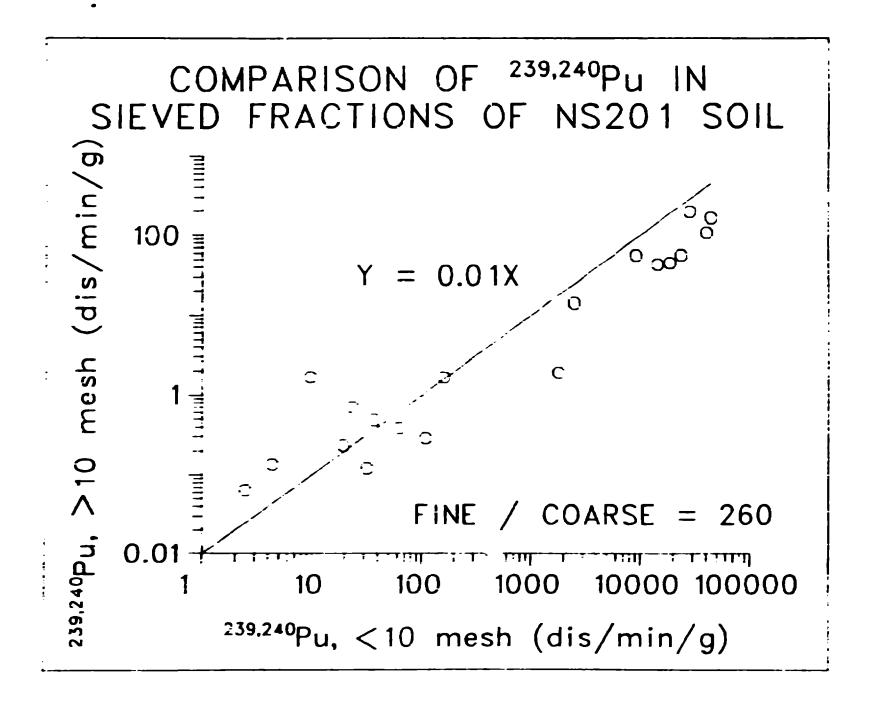
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# PLAN AND RELIEF VIEWS OF LARGE MOUND NEAR NS201 GROUND ZERO OGZ

# COMPARISON OF GAMMA DATA FOR 241Am (NS201)

	<sup>241</sup> Am, nCi/g		
LIB. NO.	LAB 1	LAB 2	
17 155	3.2	2.3	
17 157	0.43	0.29	
17164	1.6	1.3	

## COMPARISON OF GAMMA DATA FOR 137Cs (NS201)

<sup>137</sup>Cs, nCi/g

LIB. NO. LAB 1 LAB 2

17 155 0.047 0.042

17 157 0.0074 0.006 1

17 164 0.026 0.027

# COMPARISON OF GAMMA AND WET CHEMISTRY FOR 239,240Pu (NS201)

	<sup>239,240</sup> Pu, nC <sub>1</sub> /g		
LIB. NO.	GAMMA	WET CHEM.	
17 155	21.2	27.3	
17 157	4.90	4.01	
17 164	14.1	15.1 ·	

# ESTIMATED RADIOACTIVITY IN LARGE MOUND (NS201)

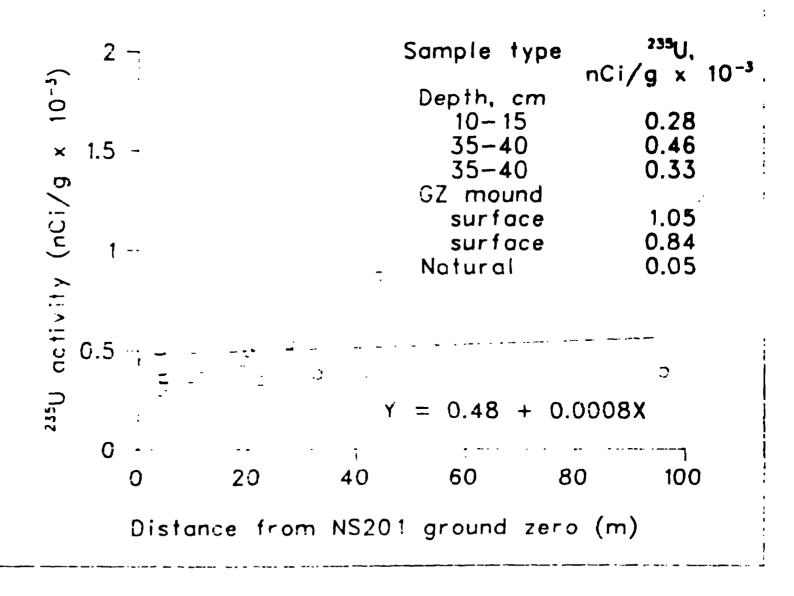
DADIONILO IDE	$A \cap T \cap T \vee$	$\cap$ :
RADIONUCLIDE	ACTIVITY,	U:

<sup>239,240</sup> Pu	7.7

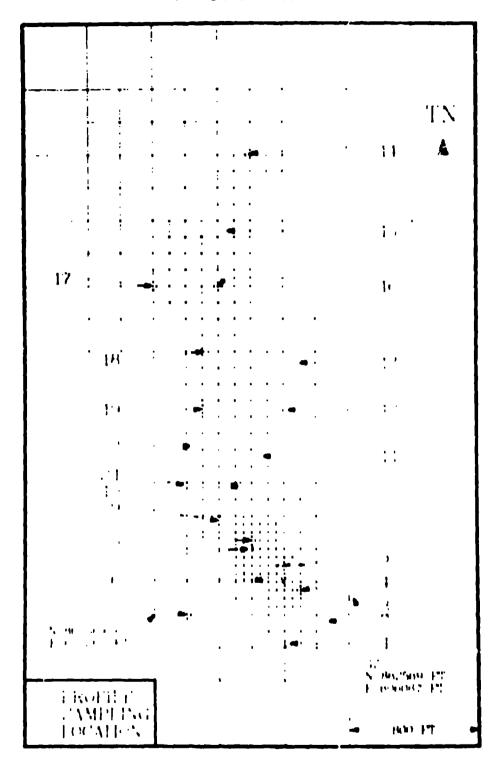
## 238U IN SOILS AT NS201

LOCA N	TION	SAMPLE TYPE	<sup>238</sup> U, nCi/g (x 10 <sup>-3</sup> )
385	275	Surface	8.6
385	175	Surface	11
335	175	Surface	11
370	400	10-15 cm	6.9
370	400	20-25 cm	4.9
-72	47	Surface mound	23
-42	40	Surface mound	4.8
		Approx. nat. <sup>238</sup> U	1.

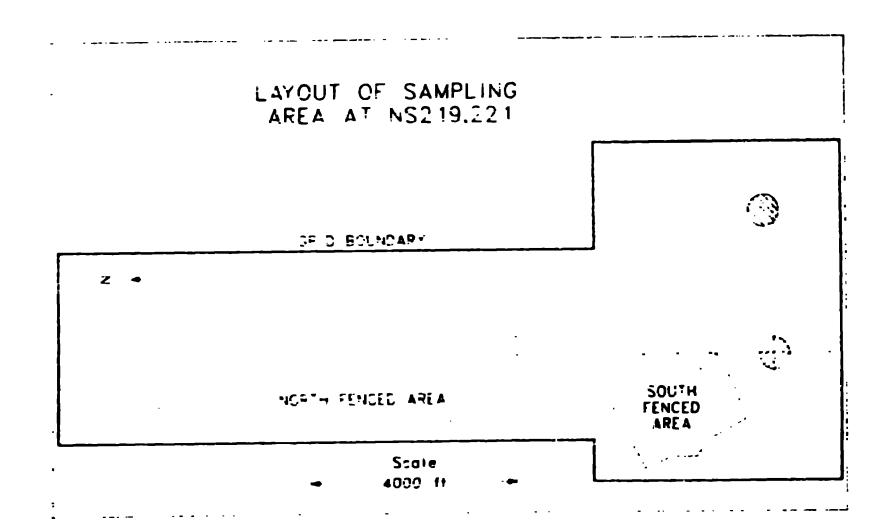
## 235U IN NS201 SOILS



## NS201 GRID



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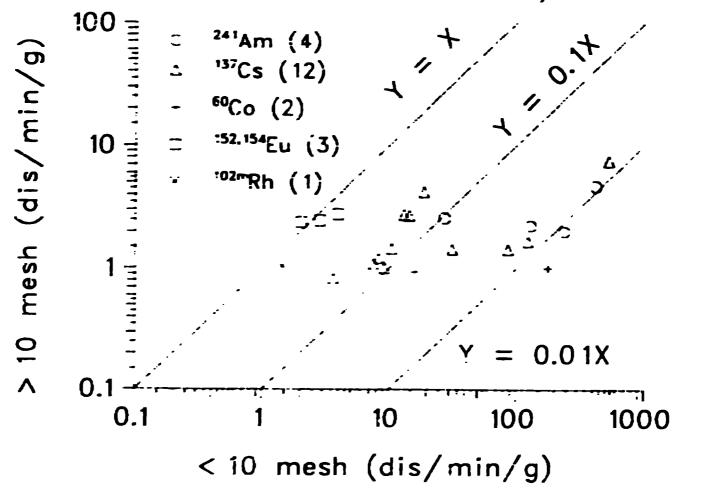
# RADIONUCLIDES IDENTIFIED IN SOILS OF NS219,221

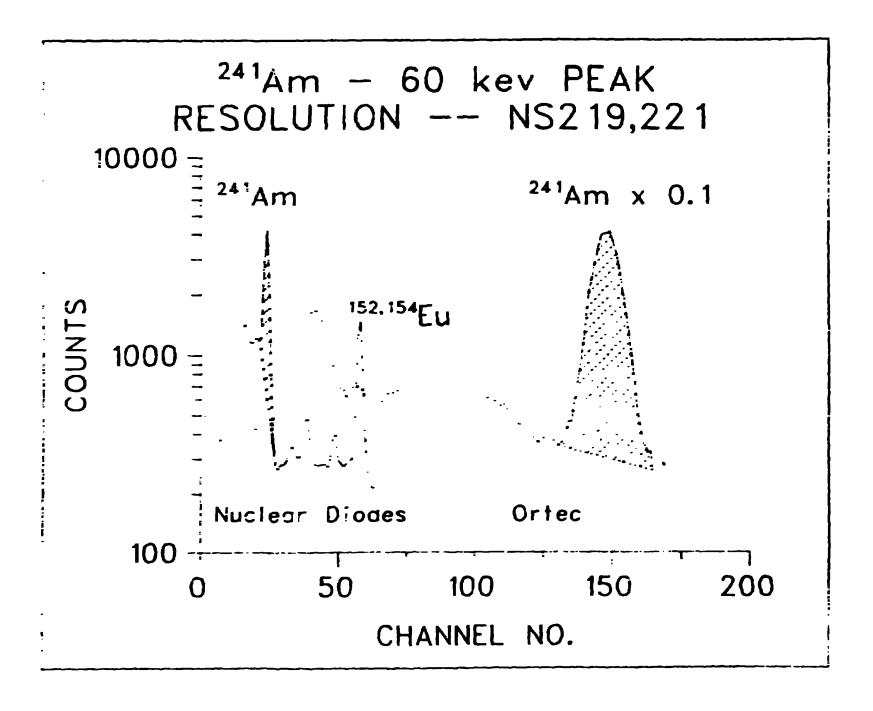
Nuclide	Decay halflife, y	Nuclide	Decay halflife, y
Alpha <sup>238</sup> Pu <sup>239,240</sup> Pu <sup>233,234</sup> U	88 2.4 x 10 <sup>4</sup> 2 x <sup>1</sup> 0 <sup>5</sup>	Gamma <sup>241</sup> Am <sup>137</sup> Cs <sup>60</sup> Co <sup>152</sup> Eu	432 30 5.3 16
CS + gamı ' <sup>25</sup> Sb	ma 2.7	<sup>154</sup> Eu <sup>155</sup> Eu <sup>102m</sup> Rh	13 1.8 2.9
LS ³H	12		

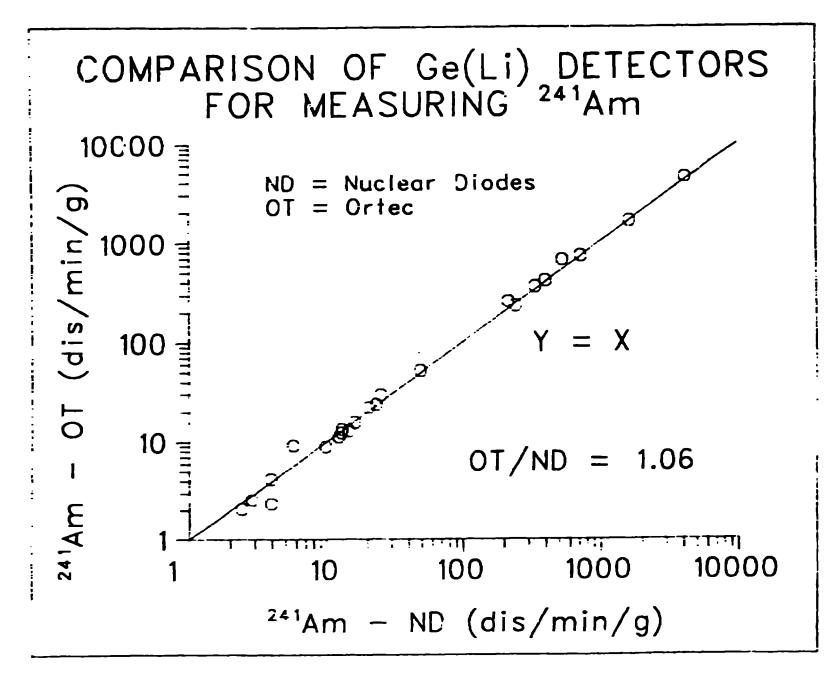
## RADIONUCLIDES LIKELY TO BE PRESENT

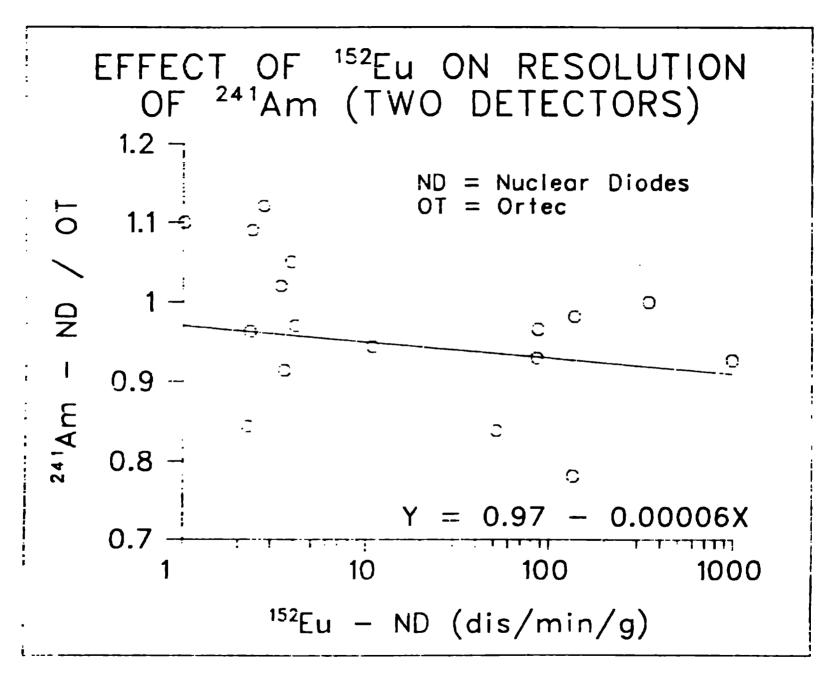
Nuclide	Decay haiflife, y	Nuclide	Decay halflife, y
<sup>90</sup> Sr - <sup>90</sup> Y	30	<sup>237</sup> Np	$2 \times 10^{6}$
' <sup>47</sup> Pm	2.6	<sup>242m</sup> Am	15 <b>Ø</b>
<sup>55</sup> Fe	2.7	•	
<sup>294</sup> Ti	5	<sup>242</sup> Cm	0.4

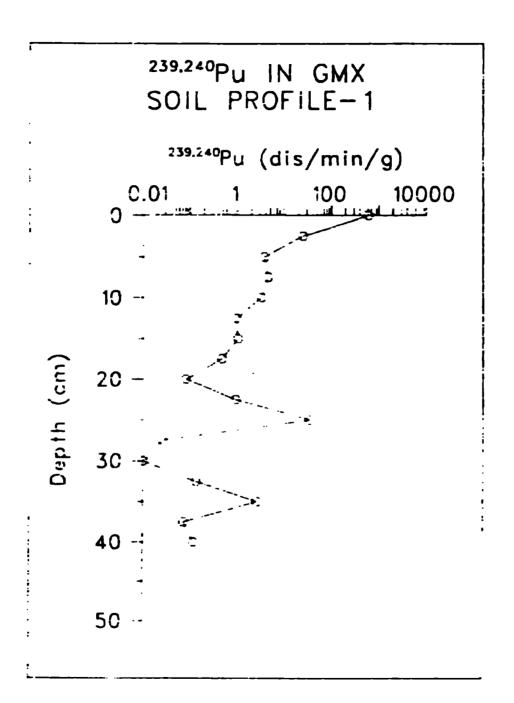
# COMPARISON OF RADIONUCLIDES IN SIEVED FRACTIONS OF NS219,221 SOIL

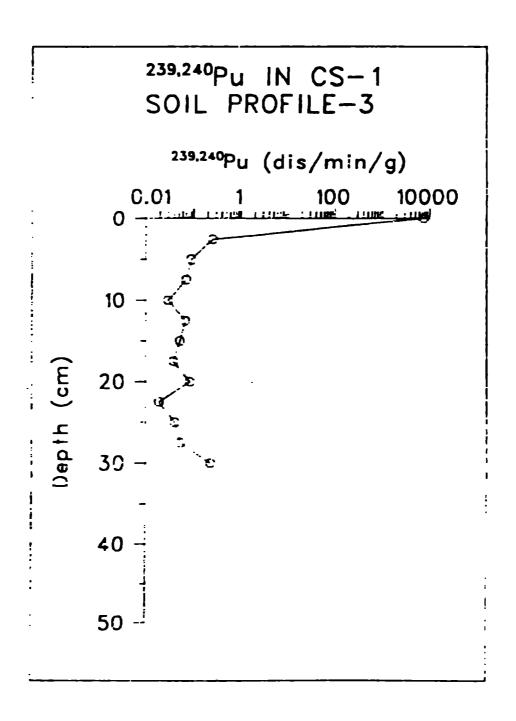


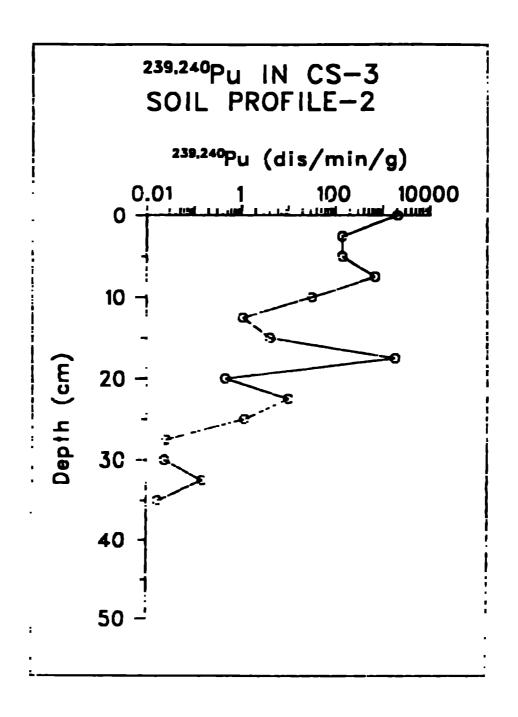












### Notes or telk

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